Semiclassical spin liquid state of easy axis Kagome antiferromagnets

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Motivated by recent experiments on Nd-langasite, we consider the effect of strong easy axis single-ion anisotropy D on $S > 3/2$ spins interacting with antiferromagnetic exchange J on the Kagome lattice. When $T \ll DS^2$, the collinear low energy states selected by the anisotropy map on to configurations of the classical Kagome lattice Ising antiferromagnet. However, the low temperature limit is quite different from the cooperative Ising paramagnet that obtains classically for $T \ll JS^2$. We find that sub-leading $\mathcal{O}(J^3S/D^2)$ multi-spin interactions arising from the transverse quantum dynamics result in a crossover from an intermediate temperature classical cooperative Ising paramagnet to a semiclassical spin liquid with distinct short-ranged correlations for $T \ll J^3S/D^2$.

Introduction: The insulating magnet Nd-langasite has localized Nd^{3+} moments that live on sites of a 2-dimensional Kagome lattice made up of corner-sharing triangles (Fig 1a). These moments carry a total angular momentum $\mathbf{J}_{\mathrm{ion}} = 9/2$ and are subject to a strong single-ion anisotropy term $D \sim 10K$ [1] that picks out the crystallographic c axis as the common easy axis of all the spins [2]. Although they interact with a sizeable nearest neighbour antiferromagnetic exchange coupling $J \sim 1.5K$ [1] (corresponding to a Curie-Weiss temperature $\Theta_{CW} = -52$K), Nd-langasite does not exhibit any magnetic order down to 50 mK [1, 3, 4]. Such behaviour contrasts strikingly to conventional insulating solids with localized magnetic moments and concomitant short-ranged exchange interactions [5]—these usually enter a magnetically ordered state at low temperature, the nature of which can often be understood quite simply in terms of the classical energetics of the leading exchange interactions.

When these interactions compete due to the geometry of the lattice, as is the case in Nd-langasite, one often obtains a large degeneracy of inequivalent classical ground states which prevents ordering and instead results in unusual cooperative paramagnetic behaviour for a range of intermediate temperatures below Θ_{CW} – such systems are called frustrated magnets [6]. Frequently, quantum effects and sub-leading interactions eventually do lead to an (often complex) ordered state at still lower temperature—Kagome lattice magnets provide many examples of this [7, 8, 9]. In some other cases, such as the $S = 1/2$ Kagome magnet *herbertsmithite* [10], there is apparently no tendency of the spins to form an ordered arrangement even at the lowest temperatures accessible to experiment.

Systems such as Nd-langasite and Herbertsmithite provide possible realizations of so-called spin-liquid states, which have been the subject of sustained theoretical activity [11] going back to the seminal work of Fazekas and Anderson [12]. Much of this theoretical activity has focused on the challenging case of Heisenberg exchange interactions (isotropic in spin space) and low spin $S = 1/2$ (in which quantum fluctuations are expected to be strongest) [11]. In the opposite Ising limit in which the exchange interactions only couple one component of neighbouring $S = 1/2$ moments on the Kagome lattice, there are no quantum effects and the system remains in a cooperative paramagnetic state with short ranged spin correlations all the way down to zero temperature [13].

In order to model the case at hand, Nd-langasite, we consider moments with larger spin $S > 3/2$ interacting with isotropic Heisenberg exchange J on the Kagome lattice, but which—unlike the case considered in [14]—are subjected to a strong single-ion anisotropy D that picks out a common easy axis for all the moments:

$$H = J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j - D \sum_i (S_i^z)^2 \qquad (1)$$

When D dominates over J, the moments prefer collinear spin states that correspond to configurations of a classical Ising antiferromagnet. However, quantum fluctuations induced by transverse components of the exchange coupling can lead to low temperature behaviour quite different from the classical cooperative Ising paramagnet, and here we explore this possibility in some detail.

Our results are readily stated: We find that such easy axis magnets *do not* develop any long-range order down to very low temperature, and are thus good examples of genuine spin-liquid behaviour in a system of quantum spins. More precisely, we find that there are two qualitatively distinct regimes separated by a cross-over temperature $T^* \sim J^3S/D^2$. For $T^* < T \ll JS^2$, the short ranged spin correlations of the system are well-described by the correlations of the cooperative Ising paramagnet described above. Below T^*, the leading effects of *virtual* quantum fluctuations begin to dominate, leading to a qualitatively different semiclassical spin liquid regime in which the liquid structure factor of the spins encodes distinctive short-ranged correlations, but there is no long range order of any kind (this is in sharp contrast to easy axis Kagome antiferromagnets with $S = 1$ moments,
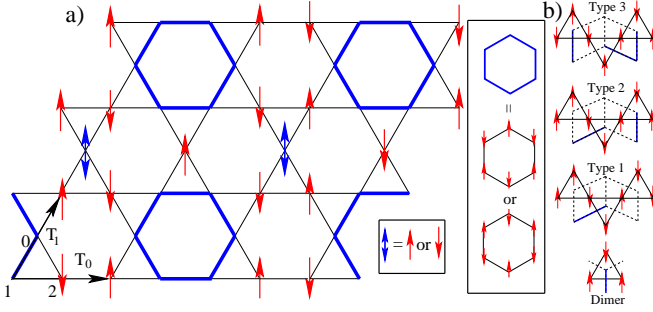
FIG. 1: (color online). a) A $L_x = 4$, $L_y = 3$ piece of the Kagome lattice, with principal directions T_0 and T_1, and three sublattices of sites marked; also shown is a class of partially ordered states with macroscopic entropy that simultaneously minimize J_1 and J_2. b) Unfrustrated bonds of type n and mapping of Ising spins to dimers on the dual dice lattice.

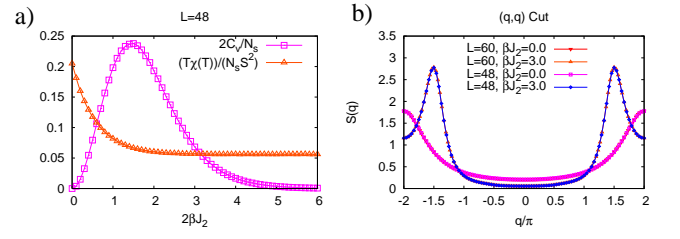
FIG. 2: (color online). (a)Specific heat C_v and uniform susceptibility χ in the crossover region. (b) The $q_x = q_y$ cut for $S(\vec{q})$ is also shown for two different system sizes ($L = 48, 60$) for two different temperatures ($\beta J_2 = 0.0, 3.0$).

where the quantum dynamics connects different classical ground states and induces spin nematic order [15]).

Effective Hamiltonian: When D dominates over J, the system prefers *collinear spin configurations* with $S_i^z = \sigma_i S$ (with $\sigma_i = \pm 1$). This degenerate ground-state manifold of the unperturbed problem can be thought of in terms of configurations of the Ising pseudo-spin variables σ. The low energy physics in this regime is then best described in terms of an effective Hamiltonian $\mathcal{H}$ that encodes the splitting of this degenerate Ising subspace to each order in J/D. To order J^3/D^2, $\mathcal{H}$ is given as [16]

$$\mathcal{H}_{\text{eff}} = J_1 \sum_{\langle ij \rangle} \sigma_i \sigma_j - J_2 \sum_{\langle ij \rangle} \frac{1 - \sigma_i \sigma_j}{2} (\sigma_i H_i + \sigma_j H_j) \quad (2)$$

where $J_1 = JS^2$, $J_2 = \frac{S^3 J^3}{4D^2(2S-1)^2}$, and the *exchange field* $H_i \equiv \Gamma_{ij} \sigma_j$ with $\Gamma_{ij} = 1$ for nearest neighbours and zero otherwise. In the above, the first term corresponds to the leading effect of the z component of the spin exchange, while the second term arises from *virtual* quantum transitions of pairs of anti-aligned spins out of the low energy Ising subspace. An additional $\mathcal{O}(J^{2S}/D^{2S-1})$ pseudo-spin exchange term, representing real quantum transitions, is subleading for $S > 3/2$ [16].

Classical Ising regime: For temperatures T well above the exchange energy scale JS^2, but lower than the anisotropy energy DS^2, the behaviour of the system will be that of to the high temperature paramagnetic regime of the classical Ising model on the Kagome lattice. As the temperature is lowered, the exchange begins to make itself felt, and the system crosses over to an intermediate temperature regime $J_2 \ll T \ll J_1$ whose physics is controlled by the ground states of the classical Kagome lattice Ising antiferromagnet (KIAF).

In these ground states each triangle has exactly one frustrated bond (connecting a pair of aligned spins). This 'minimally frustrated' ensemble of states has a residual entropy of $0.502 k_B$ per site [13], and the loss of entropy coming from the high-temperature paramagnet is reflected in a featureless peak in the specific heat C_v (the area under the C_v/T curve measuring this entropy loss).

This ensemble of ground states can be conveniently represented by dimer coverings on the dual dice lattice wherein a dimer is placed on every link of the dual lattice that intersects a frustrated Kagome lattice bond (Fig 1b); note that in this dimer model, a hard dimer constraint is operative *only* on the 3-coordinated dice lattice sites, while the 6-coordinated sites have a soft constraint that an even number of dimers touch them.

As mentioned earlier, this ensemble [13] yields correlations that are extremely short-ranged and hence a featureless spin structure factor (Fig 3a). The temperature regime $J_2 \ll T \ll J_1$ is thus a *classical cooperative Ising paramagnet* with a reduced (compared to the high temperature Ising paramagnet) value of the magnetization fluctuations as reflected in $T\chi$ (χ being the linear susceptibility to a magnetic field along the easy axis).

Minimizing J_2: To understand the behaviour of the system at still lower temperatures, we first focus on the $T = 0$ ground states of $\mathcal{H}_{\text{eff}}$ and show that minimally frustrated configurations which also minimise the multi-spin interaction J_2 satisfy the criterion that *no spin be the minority spin of both triangles to which it belongs*. To this end, we first note that the multispin interaction, projected to the minimally frustrated Ising subspace, assigns a 'potential energy' $-2J_2(n-1)$ to an unfrustrated bond of type n—here we classify an unfrustrated bond to be of type 3 (1) if the two spins it connects are both majority (minority) spins of the *other* triangles to which they belong, while an unfrustrated bond connecting one majority and one minority spin is of type 1 (Fig 1b).

As an unfrustrated type n bond of a minimally frustrated Ising configuration is surrounded by a double rhombus of the dual dice lattice with n dimers on its perimeter (Fig 1b), the projected multispin interaction J_2 translates to a dice-lattice dimer model with interactions on double-rhombii surrounding unfrustrated bonds

$$H_D = -4N_t J_2 \sum_{n=1}^{3} (n-1) f_n \quad (3)$$

where N_t is the number of triangles in the kagome lattice and f_n is the fraction of unfrustrated bonds of type n, i.e with n dimers on the perimeter of the corresponding double-plaquette (Fig 1b.).

Thus, up to a constant, the interaction energy counts the *total number* of dimers on the perimeters of all double-plaquettes corresponding to unfrustrated bonds. Since every single plaquette with n dimers on its perimeter is part of $4-n$ such double-plaquettes, the n dimers will contribute with multiplicity $4-n$ to this total number. Thus, if the fraction of single plaquettes with n dimers on their perimeter is g_n (with $\sum_{n=0}^{2} g_n = 1$, since each elementary plaquette can have 0, 1, or 2 dimers on its perimeter), the interaction energy of the configuration is $-N_P \sum_{n=0}^{2} n(4-n)g_n$, where $N_P = 3N_t/2$ is the number of elementary plaquettes. The interaction term thus assigns energies—quadratic in the number of dimers—to different types of elementary plaquettes of a dice lattice dimer configuration:

$$H_D = 2J_2 \sum_P n^2 |nP\rangle\langle nP| \qquad (4)$$

where $|nP\rangle$ denotes elementary plaquettes with n dimers on their perimeter. Since each dimer is on the perimeter of two elementary plaquettes, we have $\sum_{n=0}^{2} ng_n = 4/3$, and this constraint along with $\sum_{n=0}^{2} g_n = 1$ allows us to minimize the potential energy H_D: H_D is minimized when $g_0 = 0$ (which fixes $g_1 = 2/3$ and $g_2 = 1/3$) and this immediately gives the criterion that no spin be the minority spin of both the triangles to which it belongs.

We have investigated the set of minimally frustrated configurations that satisfy this minority spin rule in some detail, and find that it is possible to construct a large subset of states (i.e. with macroscopic entropy) satisfying this rule, and related to each other by local spin flips (Fig 1a). This construction immediately provides a lower bound of $k_B \ln(2)/6$ per site on the entropy of the ground states of H_D. The question then arises whether the correlations in the $T \rightarrow 0$ limit remain short-ranged, or whether order by disorder occurs here, and below we address this question numerically.

Loop algorithm: In order to obtain reliable numerical results that can be used to settle this delicate question, we have developed a new loop algorithm that can handle the non-trivial Boltzmann weight associated with H_D as well as keep track of the different constraints on 3- and 6-coordinated sites, while retaining the efficiency of the usual hard-core dimer model loop algorithms [17, 18].

The algorithm proceeds as follows: A change in dimer configuration is initiated by starting at a randomly chosen 6-coordinated site (say μ_0) and moving, with equal probability, to one of its six 3-coordinated neighbours, say i. If the link $\langle \mu_0 i \rangle$ is covered by a dimer, this dimer is rotated about the *pivot* site i, so that it now covers link $\langle \mu i \rangle$—μ is chosen from the three possible 6-coordinated
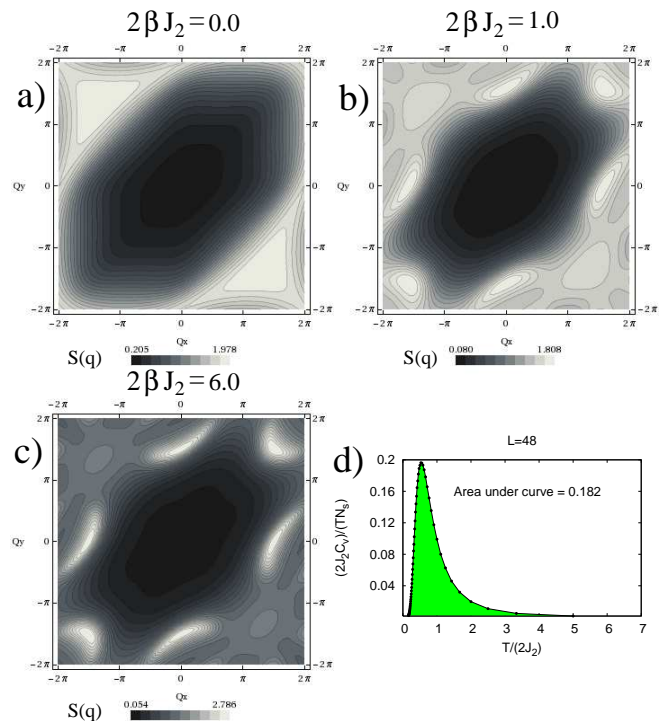

FIG. 3: (color online). (a-b-c)Spin structure factor $S(\vec{q})$ shown for three different temperatures, showing crossover to semiclassical spin liquid regime. d) Area under the C_v/T curve gives the reduction in entropy associated with the crossover to the semiclassical spin liquid.

neighbours of i according to a table of probabilities satisfying detailed balance (if $\mu = \mu_0$, the attempted update of the dimer configuration ends without making any change in the configuration). On the other hand, if the link $\langle \mu_0 i \rangle$ is unoccupied by a dimer, and the dimer touching i covers a different link $\langle \mu i \rangle$, this dimer is rotated about pivot i to cover link $\langle \mu_0 i \rangle$ with a certain probability p chosen to satisfy detailed balance (conversely, with probability $1-p$, the attempted update ends without making any change in the dimer configuration).

In both cases above, the first step of the update procedure results in a violation of the (soft) constraint at μ_0 as well as at another 6-coordinated site μ. By repeating this set of moves with probabilities chosen to satisfy detailed balance at each step, the site μ can be moved around in a closed loop until it finally meets μ_0 again and heals the 'defects' that were originally introduced at that both sites. When this happens, a large change in the dimer configuration is effected along a closed loop of links, and this large change can be accepted with unit probability, thereby providing an efficient means of sampling the Gibbs distribution associated with H_D.

The semiclassical spin liquid: From numerical simulations on $L_x = L_y = L$ size systems using this algorithm (with L ranging from 10 to 60), we see no evidence at all of any phase transition as we lower the temperature

to access the $T \to 0$ limit. This is evident from the behaviour of the specific heat per site, which converges very quickly with system size, and does not show any singularity in the thermodynamic limit (Fig 2a). In addition, the spin-spin correlators as well as the bond-energy correlators show no long range order at any wavevector down to the lowest temperatures we study (Fig 2b). The system thus remains in a short-ranged ordered spin-liquid state down to the lowest temperatures.

Although there is no phase transition, we find that the liquid state at low temperature is quite different from the intermediate temperature cooperative Ising paramagnet. This *crossover* to a distinct semiclassical spin liquid regime at low temperature (Fig 2a) is evident for instance in the temperature dependence of the specific heat per site C_v/N_s: The C_v vs T curve shows a distinct but non-singular peak at $T^* \approx 1.3J_2$ that reflects the loss of entropy during this crossover from the cooperative Ising paramagnet to the low temperature limit in which the configurations sampled predominantly obey the minimum J_2 constraint (from the area under the C_v/T curve (Fig 3d) and knowledge of the residual entropy of the cooperative Ising paramagnet, we estimate the residual entropy of the semiclassical spin liquid to be $0.32k_B$).

A clear signature of this crossover to the semiclassical spin liquid regime below T^* can be obtained by monitoring the spin structure factor (that can be probed in neutron scattering experiments) $S(\vec{q}) = |S_0(\vec{q})\exp(iq_y/2) + S_1(\vec{q}) + S_2(\vec{q})\exp(iq_x/2)|^2$ where $S_\alpha(\vec{q})$ is the Fourier transform of the spin density on sublattice α of the Kagome lattice and q_x (q_y) refers to the projection of $\vec{q}$ on to lattice direction T_0 (T_1) measured in units of inverse Bravais lattice spacing (Fig 1a). From Fig 3 a), b), c), we see that the structure factor evolves continuously from being quite featureless in the classical cooperative Ising regime $T^* \ll T \ll J_1$ to developing characteristic crescents of high intensity diffuse scattering in the low temperature semiclassical spin liquid regime $T \ll T^*$, with precursors of these features being already present at $T \sim 2T^*$. In addition, this crossover is also characterized by a change in the magnetization fluctuations as reflected in the value of $T\chi$ (Fig 2a).

Experiments on Nd-langasite: As mentioned earlier, recent experiments on the spin-9/2 easy axis Kagome antiferromagnet Nd-Langasite have seen a liquid-like state with fluctuating moments and no long range order down to $50mK$. From the estimated values [1] of the isotropic exchange interaction and the single-ion anisotropy ($J \sim 1.5K$, $D \sim 10K$), the crossover from a classical cooperative Ising paramagnet to a semiclassical spin liquid only occurs for temperatures significantly below $50mK$ ($T^* \approx 16mK$) in this model with an isotropic exchange and a simple single-ion anisotropy term (which is expected [1] to be a good starting point for Nd-Langasite).

We therefore expect a simple classical Ising description to work fairly well for the bulk of the temperature range studied in the recent low temperature experiments. Since correlations in the cooperative Ising paramagnet are extremely short-ranged and featureless, this is consistent with the fact that data from recent neutron scattering experiments (which probe the spin structure factor) show nearly featureless diffuse scattering that can be fit quite well to a model [3, 4] of spin correlations in which nearest neighbour spins are correlated, but there are no correlations of spins further away from each other.

We hope that our results provide further motivation to study this magnet at still lower temperatures, at which the crossover to the interesting low temperature semiclassical spin liquid should become apparent. Another possible avenue for exploring this crossover in greater detail involves identification of other easy axis Kagome antiferromagnets in which the separation of scales between J and D is not so large (or the overall scale of both J and D is somewhat larger) so that the crossover to the semiclassical spin liquid occurs at more easily accessible temperatures and we hope that the results of our work provide motivation for exploring this possibility as well.

[1] A. Zorko *et. al*, Phys. Rev. Lett. **100**, 147201 (2008).
[2] P. Bordet *et. al*, J. Phys: Cond. Mat. **18**, 5147 (2006).
[3] J. Robert *et. al*, Phys. Rev. Lett. **96**, 197205 (2006); .
[4] H. D. Zhou *et. al*, Phys. Rev. Lett. **99**, 236401 (2007).
[5] J. B. Goodenough, *Magnetism and the Chemical Bond* InterScience-Wiley, New York (1963).
[6] R. Moessner, Can. J. Phys. **79**, 1283 (2001).
[7] G. Lawes *et. al*, Phys. Rev. Lett. **93**, 247201 (2004); M. Kenzelmann *et. al*, Phys. Rev. B **74**, 014429 (2006).
[8] I. Hagemann *et. al*, Phys. Rev. Lett. **86**, 894 (2001); D. Bono *et. al*, Phys. Rev. Lett. **92**, 217202 (2004).
[9] K. Matan *et. al*, Phys. Rev. Lett. **96**, 247201 (2006); T. Yildrim and A. B. Harris, Phys. Rev. B **73**, 214446 (2006).
[10] J. S. Helton *et. al*, Phys. Rev. Lett. **98**, 107204 (2007).
[11] G. Misguich and C. Lhuillier in *Frustrated spin systems*, H. T. Diep (ed), World Scientific (2005).
[12] P. Fazekas and P. W. Anderson, Philos. Mag. **30**, 432 (1974).
[13] R. Liebmann *Statistical Mechanics of Periodic Frustrated Ising Systems* Springer, Berlin (1986).
[14] O. Tchernyshyov, J. Phys.: Cond. Mat. **16**, 709 (2004).
[15] K. Damle and T. Senthil, Phys. Rev. Lett. **97**, 067202 (2006).
[16] A. Sen, K. Damle, and A. Vishwanath, Phys. Rev. Lett. **100**, 097202 (2008).
[17] A. W. Sandvik and R. Moessner, Phys. Rev. B **73**, 144504 (2006).
[18] F. Alet *et. al.*, Phys. Rev. B **94**, 235702 (2005).